

Respiratory Health Hazards due to Exposure to Radon and Free Silica Among Egyptian Ceramics Workers

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Abstract: This study aimed at determining the respiratory hazards among ceramics workers exposed to silica and radon dusts. The environmental studies included measuring of respirable dust, total dust and percent of silica content. Samples from the raw materials used (talca, clay, kaolin, limestone, and feldspar) were collected and tested for activity concentration of ^{238}U - series, ^{232}Th - series and ^{40}K . This study included 150 male exposed ceramics workers and 80 male individuals as controls. The subjects were subjected to questionnaire sheet; clinical examination; plain chest X-rays; ventilatory function tests and laboratory tests. The results revealed that respiratory symptoms were more frequent among the exposed workers than the control group. There were no statistical significant differences between the two groups as regards FEV₁% and FEV₁/FVC % while FVC and VC parameters were statistically significant lower among the exposed workers than the control group. The mean percentage of the free silica was (5.2% ± 1.01) and radon progeny level was (5.67 ± 1.97 Pci/L). Tests of Radium (^{226}Ra) series, Thorium (^{232}Th) series and Potassium (^{40}K) in some raw materials showed that Feldspar represented the highest levels compared with the other raw materials. Also, the results revealed that abnormal changes in chest x-rays and decline in ventilatory functions were statistically significant among workers exposed to high free silica (more than 5%) and to radon progeny level (more than 4 Pci/L) than those exposed to free silica (less than 5%) and to radon progeny level (less than 4 Pci/L).

[Mokhtar Madkour, Nadia Badawy, Mahmoud Serri, Safia Beshir, Hisham Aziz. **Respiratory Health Hazards due to Exposure to Radon and Free Silica Among Egyptian Ceramics Workers**. Report and Opinion 2011;3(12):50-58]. (ISSN: 1553-9873). <http://www.sciencepub.net/report>

Key Words: Ceramics industry –Silica dust – Radon progeny–Pneumoconiosis-ventilatory function tests

1. Introduction

In the ceramics industry, workers are occupationally exposed to free crystalline silica in many industrial operations. It is well known that ceramics workers have elevated risk of pneumoconiosis. The prevalence of pneumoconiosis increased with the cumulative exposure (Cavariani et al., 2005). Materials commonly used in the ceramics industry containing a remarkable content of naturally occurring radionuclides. Health effects of exposure to radon progeny other than lung cancer have been of concern, including other malignancies and non-malignant respiratory diseases (Hofmann et al., 2000 and Chen et al., 2003).

Exposure to radon is the most significant element of human irradiation by natural sources. The most important mechanism of exposure is the inhalation of radon²²² (Rn^{222}) with indoor air. Concentration of Rn^{222} and its progeny are usually higher in indoor air than in outdoor air; exceptions are in tropic areas like Egypt, where Rn^{222} concentration in well-ventilated dwellings are essentially the same as in outdoor air. Rn^{222} is a noble gas produced by alpha

decay of radium²²⁶ (Ra^{226}) in the uranium decay series. Radon and its daughters are inhaled and give rise into radiation exposure of the respiratory tract (bronchial and pulmonary region). Radionuclide may enter the human body by ingestion (eating or drinking), by inhalation, or through the skin. Chronic exposure to radon in humans and animals via inhalation resulted in respiratory effects (chronic lung disease, pneumonia and fibrosis of the lung) (ATSDR, 2000).

Crystalline silica (quartz) is a ubiquitous mineral dust found worldwide. Occupational exposure to free silica occurs in several large categories of industry. The pathogenesis of silicosis begins with the inhalation of free crystalline silica particles that have favorable characteristics for deposition in the alveolar spaces. Following inhalation, the particles interact with epithelial cells and pulmonary macrophages to initiate injury and cause fibrosis (Cosio and Majo, 2002).

2. Material and methods

This study was carried out in one of the Egyptian ceramics factory located in the industrial zone of Sadat

City, El-Menofeya Governorate. There was no adequate artificial exhaust ventilation system and the natural ventilation was not adequately enough in the work-place. The studied workers did not wear any kind of protective clothing or use any respiratory protective devices during their job.

This study included 150 exposed workers (from different shifts) each shift was 8 hours/day, 5 days /week and employed in different industrial departments of the production line: Raw material and preparation, compressors, glaze, furnaces, polishing, laboratory and quality control.

The controls were matched for the same age groups and socioeconomic standards and were chosen from many departments located in the National Research Centre and all were never occupationally exposed to dust of ceramics industry and their working environment was relatively free from substantial respiratory hazards.

The exposed and the control subjects were investigated as follows:

- **The Questionnaire:** A modified British Medical Research Council questionnaire was used (Cotes, 1987).
- **Clinical examination:** Included general and local (chest) examinations
- **Plain chest X-ray:** Full sized (14" × 14" inches) roentgenograms were performed in the present study for 100 exposed workers and 80 control, and the roentgenographic findings were classified according to the I.L.O classification of pneumoconiosis (International Labor Office "I.L.O"2002).
- **Spirometric measurements** were performed using a portable spirometer (a calibrated Micro Lab 3000 series by Micro Medical 2td PLUS printer, Buckingham, UK) according to the criteria of the *American Thoracic Society (1995)*. The ventilatory function parameters in the form of forced expiratory volume in one second (FEV₁ %), forced vital capacity (FVC), FEV₁/FVC % and VC (vital capacity) were expressed as the percentage of predicted value for each person after adjustment for age, sex, race and height.
- **Laboratory Investigation:** Detection of silica in urine according to (Paul, 1960 and APHA, 1998).

Environmental Monitoring:

A) 1- Personal Respirable and Non- Respirable Dust Samples

These samples were collected using portable dust samplers. Each sampler consists mainly of a cyclone and pump cyclones were loaded with 25 mm cellulose membrane filters, pore size 0.8 μm where the respirable dust was precipitated. The non-respirable dust was collected in grit pots connected to

the cyclone base when the amount of respirable and non-respirable dust were added together to detect the total dust. The flow rates were ranged between 1.5 - 2 L /min. and sampling periods were about 30 minutes. The cyclones were fixed on the workers' clothes at the breathing zone which is the area bordered by the outside of the shoulders and from the mid- chest to the top of the head (Ness, 1991).

2- Total Dust Area Samples

These samples were collected at selected positions in the working areas using Sartorius dust sampler "Type MD8" equipped with an open face holder of 80 mm diameter, cellulose membrane filter and 0.8 μm pore size. The flow rate was 2.5 m³/h.(41.6 L/min.) and sampling periods were about 20 minutes. Filters were weighed before and after use to give the weight of dust collected. The volume of air can be calculated from the sampling flow rate and the sampling time. The dust concentration was then derived as follows:

$$\text{Dust concentration} \frac{\text{mg}}{\text{m}^3} = \frac{\text{Weight of dust collected (mg)}}{\text{Volume of air sampled (m}^3\text{)}}$$

The percentage of free silica was used to calculate the TLV_s (Threshold Limit Values) of dust containing > 1% free silica according to the formula adopted by ACGIH, (1985) as follows:

$$\text{TLV for respirable dust} = \frac{10 \text{ mg/ m}^3}{\% \text{ respirable SiO}_2 + 2}$$

$$\text{TLV for total dust} = \frac{30 \text{ mg/ m}^3}{\% \text{ respirable SiO}_2 + 3}$$

B) Radon daughters (progeny) measurements according to International Commission on Radiological Protection (ICRP, 1981).

C) Samples from the raw materials (talca, clay, kaolin, limestone, and feldspar) used were collected and tested for activity concentration of ²³⁸U - series, ²³²Th - series and ⁴⁰K according to (Bonfanti et al 1988).

Statistical Analysis

Statistical analyses were carried out using "Statistical Package for Social Science (SPSS) Inc., Chicago, IL, USA" (version 17). "Arithmetic mean" values and "standard deviations" were computed. Student's "t- test" was used for statistical evaluation to compare means of different groups. "χ² test" used to compare the qualitative data. Odds Ratio (OR) and

95% Confidence Interval (CI) were used to find the risk of occurrence of the disease among the studied groups.

3. Results

The results of this study revealed that there was no significant difference between the exposed workers and the control group regarding their age ($P > 0.05$), but there was a higher significant difference between the control (47.5%) and the exposed groups (28.7 %) as regards smoking habit ($p < 0.05$). All the respiratory tract symptoms (RTS) were more frequent among the exposed workers than the control group however, the differences were not statistically significant, except for wheezes which was statistically different ($p < 0.05$). The results of ventilatory function tests revealed that there were no statistical significant differences between the exposed and control groups as regards FEV₁% and FEV₁/FVC % ($p > 0.05$) while FVC and VC parameters were statistically significant lower among the exposed workers than the control group ($p < 0.05$). Table(1).

As regards the exposed group, the mean duration of exposure (mean \pm SD) was 9.61 ± 2.98 years and number of workers exposed for less than 10 years was 116 (77.3%) and for more than 10 years was

34(22.7%). The results environmental monitoring revealed that the mean percentage of the free silica in the factory was ($5.2\% \pm 1.01$) which exceeds the permissible limit of free silica (5%) according to the *Egyptian Environmental Law No. 4, 1994*.

The environmental measurements of radon progeny level in the factory was (5.67 ± 1.97 Pci/L) which was higher than the permissible limit reported by the Environmental Protection Agency (EPA) which was (4 Pci/L) Table (2).

The biological monitoring of urinary silica level showed a higher statistical significant difference among the ceramics workers when compared with the control group ($p < 0.05$) Table (3). The environmental measurements of Radium (²²⁶Ra) series, Thorium (²³²Th) series and Potassium (⁴⁰K) in some raw materials used in ceramics industry showed that Feldspar represents the highest levels of ²²⁶Ra, ²³²Th, ⁴⁰K (103, 63 and 490 Biquereil /Kilogram (Bq/Kg) respectively) compared with the other raw materials (Table 4).

X-ray done to exposed workers revealed that (41%) showed fibronodular shadows, (3%) showed hyper inflated chest, (7%) showed increased broncho-vascular markings (Table 5).

Table 1. Relation between exposed workers and control group as regards age, smoking habit, respiratory tract symptoms and ventilatory function tests:

	Exposed workers (150)	Control (80)	Test of Significance	P value
Age (mean \pm SD)	36.10 \pm 7.26	37.68 \pm 6.86	t- test = 1.53	>0.05
Smoking Habits	No %	No %	$\chi^2 = 10.08$	<0.05
Smokers	43 (28.7 %)	38 (47.5 %)		
Non-Smokers	86 (57.3 %)	38 (47.5 %)		
Ex-smokers	21 (14.0 %)	4 (5.0 %)		
Respiratory Symptoms	No %	No %		
Cough	59 (39.3 %)	22 (27.5%)	$\chi^2 = 0.32$	>0.05
Sputum	66 (44.0 %)	30 (37.5%)	$\chi^2 = 0.91$	>0.05
Dyspnea	46 (30.7 %)	16 (20.0%)	$\chi^2 = 3.01$	>0.05
Wheezes	54 (36.0 %)	16 (20.0%)	$\chi^2 = 6.31$	<0.05
Ventilatory Functions	mean \pmSD	mean \pmSD		
FEV ₁ %	63.1 \pm 12.3	80.6 \pm 11.1	t-test = 0.76	<0.05
FVC	68.1 \pm 8.1	86.3 \pm 7.1	t-test = 2.94	<0.05
FEV ₁ /FVC %	92.6 \pm 7.1	93.4 \pm 6.2	t-test = 0.24	>0.05
VC	65.4 \pm 4.7	87.4 \pm 7.4	t-test = 4.12	<0.05

Table(6) showed that 66.6% of workers exposed to free silica (more than 5%) had statistically and significant prevalent abnormal chest x-ray findings in the form of small fine fibro-nodular changes (s, t and p, q categories and the profusion (degree of scattering) of small opacities ranged from 1/1 to 2/1 according to ILO classification), increased broncho-vascular markings and hyper-inflated chest) in comparison to 44.3% of workers exposed to free silica (less than 5%) ($P < 0.05$). Also, this table clarified that 63% of workers exposed to free silica (more than 5%) showed decline in their ventilatory functions in comparison to 49% of workers exposed to free silica (less than 5%) with no statistical difference ($p > 0.05$).

Table (7) showed that 56% of workers exposed to radon progeny level (more than 4 Pci/L) showed higher statistical significant difference chest x-ray changes in comparison to 41.2% of workers exposed to radon progeny level (less than 4 Pci/L) ($p < 0.05$). Also, this table showed that 55.9% of workers exposed to radon progeny level (more than 4 Pci/L) showed lower statistical significant difference changes in their ventilatory functions in comparison to 50% of workers exposed to radon progeny level (less than 4 Pci/L) ($p < 0.05$).

Table 2. Airborne concentration of radon and silica

Environmental Measurements	Level
Free Silica Level (%)	5.2 ± 1.01
Radon Progeny Level (Pci / L)	6.0

Table 3. Biological monitoring of urinary silica level among the exposed and control groups

Urinary silica level U.Si/U.cr (µg/g U.cr)	Mean ± SD	t-test	P value
Exposed workers (150)	1.61 ± 1.42	2.5	<0.05
Control group(80)	1.16 ± 1.03		

U.Si/U.cr = Urinary Silica/ Urinary creatinine µg/g

U.cr = µg/g Urinary reatinine

Table 4. Level of Radium (^{226}Ra) series, Thorium (^{232}Th) series and Potassium (^{40}K) in some raw materials used:

Samples	Activity Concentration (Bq/Kg)		
	^{226}Ra - series	^{232}Th - series	^{40}K
Talc	20.17	26.54	237.88
Kaolin	10.6	5.6	32.65
Lime Stone	77.32	38.94	73.27
Feldspar	102.72	62.78	489.98
Clay	48.56	38.65	73.47

Bq/Kg = Biqurell /Kilogram

Table 5. Chest x-ray findings among the exposed workers and control subjects:

Chest x-ray findings	Groups				χ^2	P- value
	Exposed (100)		Control (80)			
	No.	%	No	%		
Normal	49	49.0%	78	97.5%	51.82	0.001
Fibro-nodular shadows	41	41.0%	0	0%		
Hyper inflated chest	3	3.0%	0	0%		
Increase broncho-vascular markings	7	7.0%	2	2.5%		

Table 6. Odds ratio of silica and risk of airway obstruction:

	Level of free silica (%)				Test of significance	P- Value	O.R (95% CI)
	$\leq 5\%$		$>5\%$				
	NO (70)	%	NO (30)	%			
Abnormal chest x-ray findings	31	44.3	20	66.6	$\chi^2 = 4.21$	<0.05	3.06 (1.12-7.94)
Abnormal Ventilatory Findings	51	49.0	29	63.0	$\chi^2 = 0.17$	>0.05	1.7 (0.85-3.54)

OR= Odds Ratio 95 % C.I = 95% confidence interval

Table 7. Odds ratio of radon and risk of airway obstruction:

	Level of radon progeny (Pci/L)				Test of significance	P- Value	O.R (95% CI)
	≤ 4 Pci/L		>4 Pci/L				
	NO (34)	%	NO (34)	%			
Abnormal chest x-ray findings	14	41.2	37	56.0	$\chi^2 = 1.99$	>0.05	2.02 (0.8-4.08)
Abnormal Ventilatory Findings	24	50.0	56	55.9	$\chi^2 = 0.32$	>0.05	1.2 (0.61-2.42)

OR= Odds Ratio 95 % C.I = 95% confidence interval Pci / L = Picocurie / Liter

4. Discussion

In the ceramic industry, workers are occupationally exposed to free crystalline silica dust in many industrial operations. It is well known that ceramic workers have elevated risk of chronic silicosis. The prevalence of silicosis increased with the cumulative exposure (Cavariani et al., 2005). Materials commonly used in the ceramic industry contain a remarkable content of naturally occurring radionuclides. Health effects of exposure to radon progeny other than lung cancer have been of concern, including other malignancies and non-malignant respiratory diseases (Hofmann et al., 2000, and Chen et al., 2003).

The present work was conducted to estimate the extent of exposure to inhaled dust including free silica level and radon emissions in Egyptian ceramic industry, to determine respiratory symptoms, radiographic chest abnormalities and pulmonary function decrements and to assess exposure response relationship between intensity of exposure and the degree of health impairment.

The collected data from questionnaire, clinical examination and ventilator function tests (VFT_s) revealed that the respiratory tract symptoms were more frequent among the exposed workers than the control group however, the differences were not statistically significant, except for wheezes which was statistically different ($p < 0.05$). This was in agreement with the results reported by several studies in the ceramic industry (Cowie et al., 2001, Sakar et al., 2005 and Saad et al., 2006).

In the present study, the incidence of the respiratory tract symptoms (except cough) were more frequent with increasing the duration of employment among ceramic workers (more than 10 years). This agree with a study done on workers exposed to crystalline silica in gold mines, and showed that the prevalence of respiratory symptoms increase with increasing the duration of crystalline silica exposure (Cowie and Mabena, 1991).

Regarding smoking habits among the studied groups, almost the respiratory tract symptoms were more frequent in the exposed ceramic workers than the control regardless their smoking habits. This strengthens the fact that the environmental exposure has a great effect on the prevalence of respiratory tract symptoms. Also, the results of this study revealed that, smoker and ex-smoker ceramic workers showed more frequent respiratory symptoms than those who never smoked. This can be attributed to the synergistic effect of smoking and environmental exposure on respiratory tract. This agree with a study done among workers exposed to crystalline silica in a brick manufacturing factory, and found that workers who smoked had more prevalent respiratory symptoms than those who never smoked (Xiao and David, 2000). This was in

agreement with Kreiss et al., (1989) who stated that the effect of silica dust on lung function pattern is different in smokers compared to non-smokers. In non-smokers, the decrease in residual lung capacity measurements is suggestive of restrictive impairment.

In the present study, The results of ventilatory function tests revealed that there were no statistical significant differences between the exposed and control groups as regards forced expiratory volume in one second (FEV₁%) and FEV₁/FVC % as an indication for pulmonary obstruction ($p > 0.05$) while forced vital capacity (FVC) and VC (vital capacity) parameters as an indication for pulmonary restriction were statistically significant lower among the exposed workers than the control group ($p < 0.05$).

Castranov (2000) stated that depending on the exposure pattern and individual susceptibility, there can be pathological states with opposing effects on pulmonary function such as chronic bronchitis, bronchiolitis and emphysema which cause airflow obstruction and fibrosis leading to primarily restrictive changes. Also our results agreed with Cavariani et al. (2005) who found relation between ceramics industry exposure and changes in the ventilatory function (restriction or obstruction) among workers. However, in Saad et al. (2006) study, the ventilatory function parameters of the ceramic workers were within the normal predicted values.

Smoking is recognized widely as a major risk factor for respiratory impairment of dust-exposed workers. Our study showed that the abnormal ventilatory functions were significantly higher among silica exposed ceramics workers than the control regardless their smoking habits. This revealed that the occupational environment play a role on ventilatory functions. This was in agreement with Hnizdo (1992) study which was made among South African gold miners exposed to silica dust. The author found that there were decline changes of the ventilatory functions in both smokers and non-smokers. Also, Bagatin et al. (1991) study on workers in the ceramic industry found that smokers had a worse ventilatory lung functions when compared to the non-smokers.

From this study it is clear that (63%) of the studied ceramic workers exposed to free silica level above 5%, showed changes in their ventilatory functions (obstruction or restriction), with an odds ratio (OR) of 1.7 (Table 5-a). Also, 54.9% of this studied group who were exposed to radon progeny level above 4 Pci/L, showed changes in their ventilatory functions.

From this study, it is clear that (66.6%) of the studied ceramic workers exposed to free silica level above 5 %, significantly showed changes in their x-ray films ($P < 0.05$). Also, 56% of this studied group who

were exposed to radon progeny level above 4 Pci/L, showed changes in their x-ray films.

The results of the environmental monitoring in this study revealed that the mean percentage of the free silica in the factory was $(5.2\% \pm 1.01)$ which exceeds the permissible limit of free silica (5%) according to the Egyptian Environmental Law No. 4, 1994. This mean concentration of free silica was lower than that measured in another study done among workers exposed to respirable crystalline silica at Chinese pottery factory which was (12 %) (Zhuang et al., 2001).

The environmental measurements of radon progeny level in the factory was (5.67 ± 1.97) Pci/L which was higher than the permissible limit reported by the Environmental Protection Agency (EPA) which was (4 Pci/L).

Rushon (2007) stated that, there are many factors influencing the variation from industry to industry in risks associated with exposure to silica-containing dusts include the presence of other minerals in the dust, particularly when associated with clay minerals; the size of the particles and percentage of quartz and the physiochemical characteristics of dust particles.

In the present study, the mean urinary silica level was significantly higher among the exposed ceramic workers [1.61 ± 1.42 U.Si/U.cr ($\mu\text{g/g U.cr}$)] when compared to the control [1.16 ± 1.03 U.Si/U.cr ($\mu\text{g/g U.cr}$)]. This result was confirmed by findings of EL-Safty et al. (2003) study who found that urinary silica median among silica exposed industrial workers was 1.66 U.Si/U.cr ($\mu\text{g/g U.cr}$) and among the control was 0.89 U.Si/U.cr ($\mu\text{g/g U.cr}$).

All building raw materials derived from rock and soil, such as clay, kaolin and limestone contain natural radionuclides, including uranium (^{238}U) and thorium (^{232}Th) and their decay products, and the radioactive isotope of potassium (^{40}K). The naturally occurring radionuclides in the building materials contribute to radiation exposure, which can be divided into external and internal exposure. External exposure is caused by direct gamma radiation, while internal exposure is caused by the inhalation of the radioactive inert gas radon (^{222}Rn , a daughter product of ^{226}Ra) and its short-lived secondary decay products (Tufail et al., 2007 and Somlai et al., 2008).

Due to complex physical, chemical and mineralogical characteristics, clays are used mainly in the manufacture of ceramics (porcelain, and wall tiles and pottery). Kaolin, commonly referred to as china clay that usually contains 85-95 % of kaolinite. Kaolin is widely used as a raw material in ceramics industry.

Measurements of levels of Radium (^{226}Ra), Thorium (^{232}Th), and Potassium (^{40}K) in some raw materials used in the production lines of the ceramics industry in Prima Factory in this study revealed that,

feldspar showed the highest levels of radionuclides compared to the other raw materials (102.72, 62.78, 489.98 Bq/kg respectively).

However, the measurements of Kaolin done in a study in Serbia in 1990/1995 in raw materials used in ceramics were higher than our study results (94.7, 72.7, 67.3 Bq/kg respectively), (Popovic and Todorovic, 2006). Also the measurements done in another study in Turkey on raw materials used in building materials were higher than our study results with mean (82, 94.8, 463.6 Bq/kg respectively) (Şeref, 2009).

The levels in the Lime stone in the present study were (77.32, 38.94, 73.27 Bq/kg respectively). In Serbia study in 1990/1995 which was done to measure the activity of natural radionuclides in building materials the levels were lower than our findings (43, 1.3, 0.45 Bq/kg respectively) (Popovic and Todorovic, 2006).

Regarding the levels in the Clay in our study were (48.56, 38.65, 73.47 Bq/kg respectively). However, when measured in Clay used for ceramics in Serbia (Popovic and Todorovic, 2006) in 1990/1995 the levels were (34, 63, 561 Bq/kg respectively) [2006]. While the levels in the study done in Turkey on raw materials used in building materials were with mean (39.3, 49.6 and 569 Bq/kg respectively) (Şeref, 2009).

The recommended upper level for ^{226}Ra activity concentration suggested by the European Commission is 200 Bq/Kg in building materials (Righi and Bruzzi, 2006). In our study, the activity concentration of ^{226}Ra in the raw materials used in the ceramics industry was below than the recommended upper level suggested by the European Commission.

5. Conclusion

From this study we concluded that occupational exposure to high levels of free silica >5% and radon >4 Pci/L in ceramic industry may affect the respiratory system of the exposed workers leading to x-ray chest abnormalities and decline in their ventilatory functions with short term exposure. Pre-placement and periodic examinations must be done regularly including chest x-ray and ventilatory functions assessment to diagnose any abnormalities early. Also, suitable natural and artificial ventilation in the work place are recommended, and workers must be advised to use suitable respiratory protective equipments. Environmental monitoring may be done regularly in order not to pass the permissible limits.

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11/21/2011